Fuel 156 (2015) 190–197

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Modes of occurrence of highly-elevated trace elements in superhigh-organic-sulfur coals

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HIGHLIGHTS

- The Guiding and Heshan coals are characterized by superhigh-organic-sulfur (SHOS).
- The SHOS coals have highly-elevated concentrations of V, Cr, Se, Mo, Cd, Re, and U.
- V, Cr, Se, Re, U, and Mo in these SHOS coals are mainly associated with organic matter.
- Cadmium is mainly distributed in sulfide minerals.
- A proportion of Re is associated carbonate fraction.

ARTICLE INFO

Article history:
Received 15 February 2015
Received in revised form 9 March 2015
Accepted 15 April 2015
Available online 23 April 2015

Keywords:
Mode of occurrence
Trace elements in coal
Superhigh-organic-sulfur coal
Late Permian

ABSTRACT

The concentrations and modes of occurrence of highly-elevated trace elements including V, Cr, Se, Mo, Cd, Re, and U in some late Permian coals preserved within marine carbonate successions from Southwest China, were investigated using inductively coupled-plasma mass spectrometry (ICP-MS), sequential chemical extraction procedures (SCEP), field emission-scanning electron microscopy in conjunction with an energy-dispersive X-ray spectrometer (FE SEM–EDS), and X-ray powder diffraction analysis (XRD). The coals present in this study are characterized by superhigh-organic-sulfur, ranging from 5.01% to 9.87%, and by highly-elevated concentrations of V (859 ppm on average), Cr (370 ppm), Se (29.3 ppm), Mo (364 ppm), Cd (3.87 ppm), Re (0.47 ppm), and U (214 ppm). The minerals in the coals are predominantly composed of illite or mixed-layer illite/smectite, which, together with quartz, were derived from sediment-source region. The SCEP results showed that elements V, Cr, Se, Re, U, and Mo are mainly associated with organic matter of the coal, and to a lesser extent, are associated with illite or mixed-layer illite/smectite. Traces of U-bearing minerals (coffinite and brannerite) were identified in the coal. Cadmium is mainly and Cr and Mo are partially distributed in sulfide minerals. A significant proportion of Re is also associated with the carbonate fraction.

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1. Introduction

Guizhou and Yunnan provinces, located in Southwest China, contain major coal resources (mainly of late Permian age) for South China (Fig. 1A). A number of previous studies have shown that some toxic trace elements, e.g., Be, F, As, Hg, and Tl, are significantly enriched in the late Permian coals in this area [1–6]. In particular, some late Permian coals intercalated with marine carbonate rocks have extremely-high organic S concentrations [7–11], usually in the range of 4 to 11%, and, thus, are classified as superhigh-organic-sulfur (SHOS) coals [12,13]. These SHOS coals are mainly located in Yanshan (Yunnan Province) and Guiding (Guizhou Province) Coalfields, and to a lesser extent, in Heshan (Guangxi Province) and Chenxi (Hunan province) Coalfields (Fig. 1) [4,11]. Some environmentally-sensitive trace elements, including V, Cr, Se, Mo, Cd, Re, and U, are highly enriched in these SHOS coals [4,8–11] but concentrations of other trace elements are generally close to the averages for the world hard coals reported by Ketris and Yudovich [14] and for Chinese coals reported by Dai et al. [5]. These highly-elevated trace elements were predominantly derived from hydrothermal solutions and then were deposited in an euxinic environment during peat stage [9,11]. The fly ash derived from combustion

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http://dx.doi.org/10.1016/j.fuel.2015.04.034
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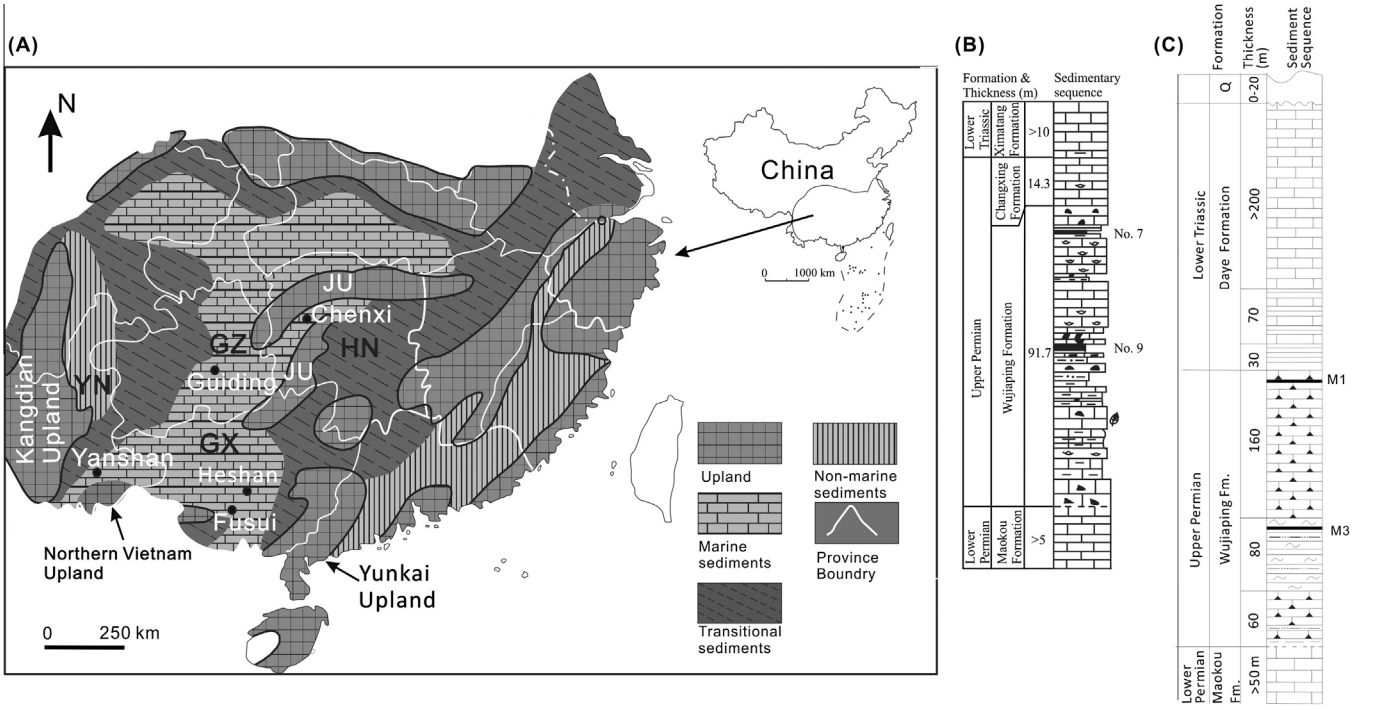

Fig. 1. Locations of Guiding and Yanshan Coalfields in southwestern China and generalized sedimentary sequences. (A) Paleoenvironment and location of the Guiding and Yanshan Coalfields [11]; (B) sedimentary sequences of the Yanshan Coalfield [9]; (C) sedimentary sequences of the Guiding Coalfield [11]. GZ, Guizhou Province; GX, Guangxi Province; YN, Yunnan Province; HN, Hunan Province. Data compiled from [9,11,26].

of these SHOS coals may also have potential economic significance for rare metals U, Se, Mo, Re, and V [11,15]. However, the modes of occurrence of these highly-elevated trace elements still remain unclear although some indirect methods (e.g., statistical analysis between ash yield and trace-element concentrations) have been employed to infer the hosts of these elements [11]. For example, based on correlation coefficients between ash yield and trace elements, Dai et al. [11] suggested that U and Mo are mainly associated with the coals' organic matter; V has a mixture of both organic and inorganic modes of occurrence; Se and Re, however, has an inorganic association in the Guiding coals. Shao et al. [8] also showed that U and Mo in the Heshan coals have an organic affinity. In this study, the modes of occurrence of highly-elevated V, Cr, Se, Mo, Cd, Re, and U in the late Permian SHOS coals from Yanshan and Guiding Coalfields were studied using sequential chemical extraction procedures (SCEP), a powerful technique that some researchers have made significant use of modes of occurrence of trace elements in coal [16–23]. The reason that we restrict the number of elements considered in this study is because of previous findings [4,8–11] showing that the concentrations of these trace elements in these coals are extremely high.

2. Background of the SHOS coals

The Yanshan and Guiding Coalfields are located in southeastern Yunnan and in the middle of Guizhou Provinces (Fig. 1A), respectively. The sedimentary sequences in the Guiding Coalfield (Fig. 1B) include the early-Permian Maokou Formation, late-Permian Wujiaping Formation, early-Triassic Daye Formation, and Quaternary system [11]. The coal-bearing strata in the Yanshan Coalfield include the Changxing and Wujiaping Formations (Fig. 1C), both of which are of the late Permian age [9]. The Changxing Formation is overlain by the early-Triassic Ximatang Formation, which does not contain coal seams. The early-Permian Maokou Formation disconformably underlies the Wujiaping Formation.

The SHOS coal (No. M9) in the Yanshan Coalfield has an average thickness of 1.91 m [9]. Two SHOS coal beds (Nos. M1 and M3), with an average thickness of 0.20 and 0.90 m respectively [11], were respectively located in the upper and middle Wujiaping Formation in the Guiding Coalfield. The coals in the Yanshan and Guiding Coalfields were deposited in tidal-flat environments of a restricted carbonate platform (Fig. 1A), and hence were preserved within marine carbonate successions. The roof sediments of the coals present in this study were mainly composed of limestones, e.g., pure limestone, flint-containing limestone, bioclastic limestone, or silicified limestone [9,11]. In a few cases, thin-layered chert or mudstone is intercalated between the coal and the roof strata. The petrological composition of the floor strata mostly consists of limestone, but in some cases of chert, marl, or mudstone [11].

The sediment-source regions, which have provided the materials of terrigenous origin during peat accumulation, are different for the SHOS coals in the two coalfields in the present study. The northern Vietnam Upland and the Kangdian Upland were respectively the terrigenous sources for the Yanshan and Guiding coals [9,11]. The two terrigenous regions have different lithological compositions, e.g., the northern Vietnam Upland mainly consisting of rhyolite [24,25] and the Kangdian Upland being dominated by mafic basalts [26].

3. Methods

Four channel samples of coal seams were collected from the mine faces, including one channel sample from the Yanshan Coalfield (sample YS-GH-9 of the M9 Coal) and three samples from the Guiding Coalfield (sample GD-GC-1 of the M1 Coal; samples GD-HST-3 and GD-GC-3 of the M3 Coal). Each coal channel sample represents an area of 10-cm wide and 10-cm deep.

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